REMARKS/ARGUMENTS

Reconsideration of this application is requested. Claims 1-15 remain active in the application subsequent to entry of this Amendment.

The Abstract

The Abstract has been revised, shortened and presented as a single paragraph to agree with current USPTO requirements.

Discussion of Amendments to the Claims

Claim 1 has been amended in order to more particularly point out and distinctly claim that which applicants regard as their invention. The process being claimed is a continuous one and this has been emphasized in two separate parts of claim 1.

In addition, claim 1 has been amended to specify that the lower value of the BM/TM ratio in formula (I) is 2.1. Basis for this appears in the description of the invention and in particular page 23, lines 14-15 of the specification. This feature serves to further distinguish the claims from the prior art as discussed in the remarks that follow.

One of the characterizing features of present invention is maintaining the BM/TM ratio in formula (I) at at least 2.1 as defined in amended claim 1. As described in the BACKGROUND ARTS portion of the present specification, in all of the conventional direct continuous polymerization methods for producing the polybutylene terephthalate, it has been difficult to prevent deactivation of the titanium compound catalyst while simultaneously decreasing the amount of the by-produced tetrahydrifuran in the early esterification reaction stage.

The present invention is directed to solving these conventional problems. An object of the present invention is to provide a process for producing polybutylene terephthalate (PBT) having excellent color tone, transparency and quality stability as well as a reduced content of impurities while preventing the loss of raw materials (*see* the discussion at page 8, lines 2 to 7 of the present specification).

Applicants have found that when the esterification reaction between terephthalic acid (TPA) and 1,4-butanediol (BG) is conducted in the presence of a titanium compound catalyst having a specific concentration under a specific pressure at a specific molar ratio between the raw materials, the amount of the titanium compound catalyst deactivated can be remarkably lessened, thereby readily overcoming the above conventional problems. The present invention

has been attained on the basis of the above finding (*see* page 8, lines 9 to 18 of the present specification).

Particular attention is directed to the formulae (I) and (II) which define optimum conditions for the esterification reaction vessel, which conditions are required to prevent deactivation of the titanium compound catalyst in the esterification reaction, and minimize the amount of by-produced THF in the process for producing PBT by continuously esterifying terephthalic acid and 1,4-butanediol as raw materials with each other. The formulae (I) and (II) are established on the basis of the finding that the deactivation of the titanium compound catalyst is more effectively prevented, the lower the concentration of the titanium compound used in the esterification reaction, the higher is the concentration of 1,4-butanediol and the lower the water concentration (*see* page 21, lines 11 to 23 of the present specification).

In the present invention the number of moles (TM) of terephthalic acid supplied per unit time means the number of moles of terephthalic acid supplied as a raw material usually in the form of a slurry mixture with 1,4-butanediol, and is generally proportional to the amount of PBT produced per unit time (*see* page 21, line 44 to page 22, line 3 of the present specification).

From the above, "BM/TM (molar ratio)", "the amount of the titanium compound catalyst", "the pressure in the esterification" affect to the amount of by-produced THF and the deactivation of the titanium compound catalyst (generation of impurities). However, these factors are not independent of each other -- these factors relate to each other mutually and affect each other. In the present invention, the specific ranges where the amount of by-produced THF and the generation of impurities are reduced, are defined by the above formulas (I) and (II).

More specifically, as seen from comparing Examples 1 to 4 and Comparative Examples 1 to 4, when the β value in the formulas (I) and (II) is within the range of 4.0 to 7.0, one will obtain a PBT resin having excellent color tone and transparency as well as a reduced impurity content (*see* page 59, Table 1, page 60, Table 2 of the present specification).

The present invention is based upon the above technical concept as reflected in the claims and has a high industrial value in a commercial process for producing polybutylene terephthalate.

<u>Response to Rejections of claims 1-8, 11-12 and 15 (Obviousness over Iida et al US 4,656,241)</u>

At the outset one must bear in mind that Iida relates to a process for producing PBT using a batch process. At column 3, lines 42- to 43 Iida states that "The present invention is basically

1431603

practiced according to a batch process." Also, all of Examples 1-11 of Iida are conducted by use of a batch process.

The batch process is completely different from the continuous process. In the batch process, at the reaction starting stage, BG and TPA are mixed, and as the reaction proceeds, the content of oligomer generated in the reaction is increased, and finally, the BG and TPA are consumed. Therefore, the composition is changed with the passage of time. So too is the reaction temperature, with the passage of time, as the temperature is raised from a low temperature of about 150°C to a high temperature of about 230°C. On the other hand, in the continuous process, the BG, TPA and oligomer are constantly present in the esterification reactor, and BG and TPA are directly fed into the high temperature reaction field of about 230°C. Therefore, the conversion of BG to THF is accelerated and the amount of by-produced THF is increased. Further, since the time when catalyst is subjected to high temperature is long, the amount of deactivated catalyst is increased.

As seen from the above, since the batch process and continuous process are completely different in starting materials, products, behavior of by-products, temperature conditions and the like, it is impossible to ascertain the optimum reaction conditions in a continuous process from the reaction conditions in a batch process.

The Examiner states that "the reference teaches ranges of pressure (500 mmHg=66.7kPa), BM/TM ratio (1.6), α (22.7 ppm) and resultant β (3.92) that overlaps the claimed range" referring to Example 8 of Iida. This is not a valid comparison for the reasons explained below.

The BM/TM ratio defined in Iida is the total amount of BG to TPA (molar ratio) used, namely it is merely the molar ratio of starting materials used for the reaction. On the other hand, in the present invention, as seen from the above, "TM is the number of moles of terephthalic acid supplied to the esterification reaction vessels per unit time; BM is the number of moles of whole 1,4-butanediol supplied <u>continuously</u> to the esterification reaction vessels per unit time" as reflected in applicants' claims. Therefore, the definition of the BM/TM ratio is completely different as between Iida and the present invention. This is not a basis for a proper comparison.

The Examiner recognizes that the BM/TM ratio in the present invention is the molar ratio of starting materials and attempts to calculate the β value. However, since the technical

conception of BM/TM ratio of Iida is completely different from that of the present invention, it is impossible to derive the β value parameter of present invention from the information in Iida. Even if the BM/TM ratio of present invention is the molar ratio of starting materials, the BM/TM ratio of Example 8 in Iida is 1.6 and lower than the lower limit of the BM/TM ratio of the present invention of not less than 2.1 as stated in amended claim 1.

Further, the Examiner states that the concentration (unit: ppm) of the titanium catalyst (α) in Example 8 of Iida is 22.7 ppm.

In example 8, as the material TPA, "755 parts of TPA containing 0.08% of acetic acid" was used and as the titanium catalyst, "di-n-butoxybis(triethanolaminate) titanium solution in n-butanol" was used. Since 60 parts (0.08%) of TPA corresponds to acetic acid, the amount of TPA as the reactive material was 695 parts.

Since the PBT unit was produced in the molar amount equal to the TPA molar amount, the produced PBT amount is calculated as follows:

695 parts/166 (molecular weight of TPA) X 220 (molecular weight of PBT unit) = 921 parts

Since the molecular weight of Ti catalyst used is 490, the Ti amount is calculated as follows:

0.75 X 0.75/490 X 48 = 0.055 parts

Therefore, the Ti concentration (α) in the PBT is calculated as:

0.055/921 = 59.7 ppm

As explained in the above, the definition of BM/TM is completely different between Iida and the present invention. However, if the β value is calculated according to the Examiner's statement, β value is calculated as follows:

$\beta = 1.6 - 0.03 \times 59.7 + 0.045 \times 66.7 = 2.8$

From the above, if the concept of the BM/TM ratio in Iida is assumed to be equal to the BM/TM ratio of the present invention, the calculated β value of Example 8 in Iida is 2.8 and this value is completely out of the scope of the present invention (β = 4.0 to 7.0) as stated in claim 1. More specifically, the reaction conditions are completely different between the batch process and a continuous process, and as such it is impossible to adapt the conditions of formulas (I) and (II) to the batch process of Iida. Namely, the above discussion and data/calculations show that the

reaction conditions in the present invention (continuous process) cannot be derived from the batch process of Iida.

As discussed above, since the batch process in Iida is completely different from the continuous process according to the present invention in the production process, there is no reasonable suggestion of the features and technical merits of the present invention. Therefore, the inventions of claims 1 to 8, 11 to 12 and 15 are not "obvious" from Iida.

Response to Rejection of Claims 9-10 and 13 (Obviousness from Iida et al. (US 4,656,241) in view of Heinze et al (US 4,499,261) and Hoeschele (US 3,801,547))

In Heinze's continuous process of producing PBT, the recycled 1,4-BG is 1,4-BG distilled from the polymerization reactor (*see* (a) of claim 1).

On the other hand, in the invention of present claim 9, the recycled 1,4-BG is 1,4-BG distilled from the esterification reactor (*see* items (2) and (3) of claim 9).

Further, in Heinze, the vapor from the polycondensation reactor 5 is condensed in the condenser 12 to separate the polymerization reaction distillate into two (lines 14 and 15), and one (line 15) is recycled to column 8 to mix with the gas phase of esterification reaction mixture, and the other (line 14) is recycled to the first transesterification reactor 1 (*see* column 3, lines 52 to 60 and Fig. 1). Namely, the recycled 1,4-BG is the 1,4-BG distilled from the polymerization reaction column and a part of it is recycled to the gas phase at the upper portion in the esterification reactor. On the other hand, in the invention of present claim 9, as described above, the recycled 1,4-BG is the distilled from the esterification reaction vessel (*see* (3) of claim 9).

From this, the invention of Heinze is completely different from the invention of present claim 9 in the source of recycled 1,4-BG and recycled portion thereof.

Further, in Heinze, the titanium catalyst is fed into the esterification reaction vessels with the material DMT and 1,4-BG (*see* column 5, lines 3 to 7). On the other hand, the invention of present claim 9, 10% or more by weight of titanium catalyst is fed to the liquid phase of reaction mixture independently to TPA (*see* (4) of claim 9). From this, the invention of Heinze is completely different from the invention of present claim 9 in the feed of titanium catalyst.

The process for producing PBT described in Hoeschele is a batch process and not continuous process according to the present invention. (The distinctions between the batch and

continuous processes are explained above.) Hoeschele merely describes a preparation of titanium 1,4-BG solution.

As discussed above, in these three cited references, there is no description nor suggestion of the technical features and merits of present claims 9-10 and 13. Therefore, the inventions of present claims 9-10 and 13 are not "obvious" from the disclosures of these references.

Response to Rejection of Claim 14 (Obviousness from Iida et al. (US 4,656,241) in view of Borman et al. (US 4,329,444))

Iida et al is discussed in detail above.

The process for producing PBT described in Borman is a batch process and not continuous process according to the present invention. Borman merely reports that heated BG at 175 to 275°C is reacted by the addition of TPA, that is a material slurry is prepared at 175 to 275°C and reacted (*see* claim 1).

On the other hand, the temperature of 1,4-BG in the invention of present claim 14 is supplied independent of terephthalic acid and is completely different from the material slurry temperature in Borman.

Further, the Examiner asserts that "Borman further teaches the motivation of minimizing the contact time between TPA and BG". However, in the present invention, the reason for the feed of 1,4-BG independently at 150 to 90°C as defined in the present claim 9 is that "When the temperature of the separately supplied 1,4-butanediol returned to the liquid phase portion of the reaction solution is too high, the amount of THF by-produced tends to be increased. When the temperature of the separately supplied 1,4-butanediol returned to the liquid phase portion of the reaction solution is too low, thermal load tends to be increased, resulting in loss of energy." (*see* page 28, lines 6 to 16 of the present specification). Therefore, the reason for this is not merely "minimizing the contact time between TPA and BG".

Borman provides no description nor suggestion of technical features and merits as defined in the present claim 14.

As discussed above, in the above cited references, there is no description nor suggestion of technical features and merits of present claim 14. Therefore, the invention of present claim 14 is not obvious from the above cited references.

For the above reasons it is respectfully submitted that claims 1-15 define patentable and inventive subject matter. The allowance of these claims is solicited. Should the examiner require further information, please contact the undersigned.

Respectfully submitted,

NIXON & VANDERHYE P.C.

By: ______________________
Arthur R. Crawford
Reg. No. 25,327

ARC:eaw
901 North Glebe Road, 11th Floor
Arlington, VA 22203-1808
Telephone: (703) 816-4000
Facsimile: (703) 816-4100